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## Spin oscillation and its reduction in a quantum dot

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Exact spectra of two electrons in quantum dots (QD's) with parabolic potentials in a magnetic field are obtained. The spectra are dramatically changed with the variation of the field and that the spin of the ground state oscillates with magnetic field. The reason is found by studying confinement dependence of the interaction energies in detail. The lowest singlet and triplet states of two electrons in disklike QD's with fixed thickness and a positively charged ion in magnetic field are calculated by a variational method. It is found that the broken symmetry of a QD with an ion has an important effect on the spin oscillation. The ion reduces the oscillation, and the oscillation reduction and the binding energies are strongly dependent on the broken symmetry related to the dot-reflection symmetry and the ion position. The results predict a possibility to observe phenomena related to ion-electron and electron-electron interactions in broken symmetry QD's. [S0163-1829(98)07543-2]

### I. INTRODUCTION

Rapid advances in semiconductor technology have recently led to the fabrication of quantum dots (QD's).<sup>1</sup> Essentially, two-dimensional electrons are laterally confined by an artificial potential. Typical sizes of confined dots laterally are about 10–100 nm and each dot contains a few electrons.<sup>2–4</sup> Alternatively, we can consider QD's as artificial atoms where the confining potential replaces that of the nucleus.

Recently, the study of semiconductor QD's has been expanded rapidly.<sup>1–7</sup> Transport measurements have shown that tunneling of electrons traversing a few electron QD is strongly influenced by the Coulomb interaction leading to Coulomb blockade effects and single-electron tunneling.<sup>2–5</sup> Applying a magnetic field on a few electron QD can reveal various spectra since the spectra of a few electron QD in a magnetic field are governed by the interplay of three kinds of energies: the electron-electron interaction energies, the Zeeman energies, and the confinement energies associated with quantization due to the QD and field confining potentials.<sup>3–7</sup> The spectra in magnetic field can be obtained from the conductance measurements in the finite drain-source voltage regime. It has been experimentally found that a magnetic field can induce transitions between the ground and excited states in semiconductor QD's containing a few electrons.<sup>4</sup>

One of the most interesting phenomena of a few electron QD is the spin oscillation of the ground state with magnetic fields,<sup>4–6</sup> which is due to the interplay of the three kinds of energies mentioned above. Based on the exact solutions of two electrons in a QD in magnetic fields, the phenomenon is clearly shown and explained in this paper.

Negative donor ions ( $D^-$ ) represent the simplest system in which the electron-electron and ion-electron interactions are significant effects. In recent years there has been extensive theoretical and experimental interest in neutral donor ( $D^0$ ) and  $D^-$  in quantum wells<sup>8–15</sup> and QD's.<sup>16</sup> These works, however, are mainly concerned with ions situated at

the center of a square quantum well.<sup>8–13</sup> Only very recently, the effects of broken reflection symmetry, i.e., those of off-well-center ion and graded quantum well have been noted<sup>14,15</sup> and some of them have been identified experimentally.<sup>15</sup> The observation has represented a verification of the predicted magnetic-field-induced unbinding (magnetic evaporation) of shallow impurity states for the off-well-center  $D^-$ -ion system.<sup>14,15</sup>

What about  $D^-$  states in QD's in magnetic fields and how large are the broken symmetry effects? Does the positive ion influence the oscillation of two-electron ground states in QD's? In this paper, we calculate the lowest spin-singlet and spin-triplet states in QD's with broken reflection symmetry in magnetic fields and provide answers to the questions.

The organization of this paper is as follows. In Sec. II, we describe the model and Hamiltonian of the problem. In Sec. III, the procedures to obtain the exact and variational solutions are briefly outlined. We define the electron-electron interaction energies of two electrons and the binding energies of  $D^-$  states in QD's. Spectra of two electrons in QD's with magnetic fields are shown and the spin oscillation is discussed on the basis of the electron-electron interaction energies in Sec. IV. For disklike QD's with a fixed thickness, the broken symmetry effects on the binding energies of the lowest singlet and triplet  $D^-$  states are studied and the reduction effect of a positive off-dot-center ion on the oscillation is clearly shown in Sec. V, followed by a summary in Sec. VI.

### II. MODEL AND HAMILTONIAN

The motion in the  $z$  direction in experimentally created quantum dots is always frozen out into the lowest subband. The corresponding extent of the wave function is much less than the one in  $x$ - $y$  plane so that the shape of dots is disklike. For most dots, a parabolic potential is a very good approxi-

mation to describe the lateral confinement of the electrons.<sup>3,4,17,18</sup>

Consider the  $i$ th electron in such a disklike QD of a thickness  $L$  with and without a positive donor ion situated at  $D$  from the QD center in the  $z$  direction. It is reasonable to put the Hamiltonian in the form

$$H_0(i) = -\nabla_i^2 - \frac{2w}{\sqrt{\rho_i^2 + (z_i - D)^2}} + V(z_i) + \frac{1}{4} \gamma_d^2 \rho_i^2, \quad (1)$$

where  $w=1$  and  $w=0$  correspond to the QD with and without the ion center, respectively, and the effective atomic units are used. The effective Rydberg  $Ry$  and the effective Bohr radius  $a^*$  are taken to be the energy and length units, respectively. It is easy to see that the  $\gamma_d^{-1/2}$  is related to the lateral confinement region of electrons in the dot. The confinement potential in the  $z$  direction is taken to be

$$V(z) = \begin{cases} V_0 & \text{for } |z| \geq L/2, \\ R(L/2 + z) & \text{for } |z| < L/2, \end{cases} \quad (2)$$

where  $R$  is the constant gradient of the potential. The  $V_0$  and  $R$  can be determined by a fixed ratio of the band-gap discontinuity. Both of  $D$  and  $R$  are related to the broken reflection symmetry.

Applying a magnetic field  $B$  perpendicular to the  $x$ - $y$  plane, Eq. (1) is turned into

$$H(i) = H_0(i) + \frac{1}{4} \gamma_b^2 \rho_i^2 + \gamma_b L_{z_i}, \quad (3)$$

where the magnetic field  $\gamma_b$  is measured in the unit  $\hbar \omega_c / 2 Ry$  with the cyclotron frequency  $\omega_c$ .  $\gamma_b L_{z_i}$  is the Zeeman term induced by the magnetic field. It is interesting to note how large the units of semiconductor materials are. For GaAs materials, for example,  $Ry = 5.8 \text{ meV}$ ,  $a^* = 10 \text{ nm}$ , and  $\gamma_b = 1$  corresponds to  $B = 6.75 \text{ T}$ .

The Hamiltonian for two electrons with the positive ion, i.e.,  $D^-$  situated at the  $D$  in the QD with the field is given by

$$H_D = H(1) + H(2) + \frac{2}{\sqrt{\rho_{12}^2 + (z_1 - z_2)^2}}, \quad (4)$$

where  $\rho_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$ , and  $H(1)$  and  $H(2)$  correspond to  $i=1$  and  $i=2$  of Eq. (3), respectively.

Setting up  $w=0$  in the two-dimensional (2D) limit of  $V_0 \rightarrow \infty$  and  $L \rightarrow 0$ , the Hamiltonian of two electrons in a two-dimensional quantum dot (2D QD) with a magnetic field is obtained from Eq. (4) to be in the form

$$H_{2e} = -\nabla_1^2 - \nabla_2^2 + \frac{1}{4} \gamma^2 \rho_1^2 + \frac{1}{4} \gamma^2 \rho_2^2 + \frac{2}{\rho_{12}} + \gamma_b L_{z_1} + \gamma_b L_{z_2}, \quad (5)$$

where  $\gamma^2 = \gamma_d^2 + \gamma_b^2$ .

### III. EXACT AND VARIATIONAL SOLUTIONS

The Hamiltonian of Eq. (5) can be separated into center-of-mass and relative-motion terms as

$$H_{2e} = H_R + H_r, \quad (6)$$

with

$$H_R = -\frac{\nabla_R^2}{2} + \frac{1}{2} \gamma^2 R^2 + \gamma_b L_{ZR} \quad (7)$$

and

$$H_r = -2\nabla_r^2 + \frac{1}{8} \gamma^2 r^2 + \gamma_b L_{zr} + \frac{2}{r}, \quad (8)$$

where  $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ ,  $\nabla_R = \nabla_1 + \nabla_2$ ,  $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ , and  $\nabla_r = (\nabla_1 - \nabla_2)/2$ .  $L_{ZR}$  and  $L_{zr}$  are the  $Z$  and  $z$  angular momentum operators in center-of-mass and relative-motion systems, respectively. This separability and the cylindrical symmetry of the problem allow us to write the two-particle wave functions in plane polar coordinates  $\mathbf{r} = (r, \varphi)$  in the form  $\Phi(R) \phi(r) \exp(im\varphi)$ . The spatial part of the total wave function is symmetric (antisymmetric) with respect to particle permutation ( $\varphi \rightarrow \varphi + \pi$ ) for even (odd) azimuthal quantum numbers  $m$ . Since the Pauli exclusion principle requires the total wave function to be antisymmetric, we have spin singlet ( $s=0$ ) and triplet ( $s=1$ ) states for even and odd  $m$ , respectively. The energy eigenvalues of Eq. (6) are given by

$$E(N, M) = (2N + |M| + 1) \gamma + M \gamma_b \quad (9)$$

with radial ( $N=0, 1, 2, \dots$ ) and azimuthal ( $M=0, \pm 1, \pm 2, \dots$ ) quantum numbers. The eigenvalues of the relative motion excluding the electron-electron interaction are also in the same kind of form and given by

$$E_0(n, m) = (2n + |m| + 1) \gamma + m \gamma_b \quad (10)$$

with the corresponding radial and azimuthal quantum numbers  $n=0, 1, 2, \dots$ , and  $m=0, \pm 1, \pm 2, \dots$ .

However, we should solve the Schrödinger-like equation

$$H_r[\phi(r) \exp(im\varphi)] = E(m)[\phi(r) \exp(im\varphi)] \quad (11)$$

to obtain the energy of the relative motion including the electron-electron interaction. It is easy to find the equation satisfied by the function  $\phi(r)$ :

$$\frac{d^2 \phi}{dr^2} + \frac{1}{r} \frac{d\phi}{dr} + \left( \frac{E(m) - m \gamma_b}{2} - \frac{1}{r} - \frac{m^2}{r^2} - \frac{1}{16} \gamma^2 r^2 \right) \phi = 0. \quad (12)$$

Now, we are prevented from analytically exact solutions of the eigenvalue problem because Eq. (12) with suitable boundary conditions is beyond the analytical problem of confluent hypergeometric equations. However, we can use the method of series expansion<sup>19</sup> to obtain exact series forms in different regions of Eq. (12) and the exact values of  $E(m)$  and, then, the exact solutions of two-electrons in the quantum dot in a magnetic field.

In the region  $0 < r$  we have a series solution, which has a finite value at  $r=0$  as follows:

$$\phi(r) = A r^{|m|} \sum_{n=0}^{\infty} a_n r^n, \quad (13)$$

where  $A$  is a constant and  $a_0$  is equal to 1. The other  $a_n$  can be determined by the recurrence relation. In the region  $r < \infty$  we obtain a normal solution in the form

$$\phi(r) = B \exp\left(-\frac{1}{8} \gamma r^2\right) r^s \sum_{n=0}^N b_n r^{-n}, \quad (14)$$

with

$$s = [E(m) - m \gamma_b] / \gamma - 1, \quad (15)$$

where  $B$  is a constant.  $b_0 = b_1 = 1$  and the others are determined by the recurrence relation.

In order to match the solution of Eq. (13) with that of Eq. (14), we give  $T$  solutions around  $R_1, R_2, \dots$ , and  $R_T$ . The solution of uniformly convergent Taylor series around  $R_i$  ( $i = 1, 2, \dots, T$ ) is written as follows:

$$\phi(r) = C_i \sum_{n=0}^{\infty} c_{in} (r - R_i)^n + D_i \sum_{n=0}^{\infty} d_{in} (r - R_i)^n, \quad (16)$$

where  $C_i$  and  $D_i$  are constants,  $c_{i0}$  and  $d_{i1}$  are equal to 1, and  $c_{i1}$  and  $d_{i0}$  are equal to 0. The  $c_{in}$  and  $d_{in}$  can be determined by the recurrence relations.

Using the matching conditions at  $r = R_i$  ( $i = 1, 2, \dots, T$ ) and the  $2 \times 2$  transfer matrices, we deduce the equation for eigenenergies  $E(n, m)$  easily. The values of  $E(n, m)$  and  $\phi_{nm}(r)$  are obtained numerically. For the sake of convenience, we define the electron-electron interaction energies  $E_r(n, m)$  as the difference between  $E(n, m)$  and  $E_0(n, m)$ , i.e.,

$$E_r(n, m) = E(n, m) - E_0(n, m). \quad (17)$$

Then, the energy eigenvalues of the Hamiltonian in Eq. (5) are the sum of  $E(n, m)$  and  $E(N, M)$  as follows:

$$E(n, m; N, m) = [2(N + n) + |M| + |m| + 2] \gamma + [M + m] \gamma_b + E_r(n, m). \quad (18)$$

There are no exact solutions for Eqs. (1) and (4). Therefore approximation methods should be used. To solve the problem, we rewrite Eq. (1) as

$$H_0(i) = H_0(\lambda_i, \alpha_i, \rho_i, \phi_i) + H_0(z_i) + H'(\lambda_i, \alpha_i) \quad (19)$$

with

$$H_0(\lambda_i, \alpha_i, \rho_i, \phi_i) = -\frac{1}{\rho_i} \frac{\partial}{\partial \rho_i} \left( \rho_i \frac{\partial}{\partial \rho_i} \right) - \frac{1}{\rho_i^2} \frac{\partial^2}{\partial \phi_i^2} - \frac{2\lambda_i}{\rho_i + \alpha_i} + \frac{1}{4} \gamma^2 \rho_i^2 + \gamma_b L_{z_i}, \quad (20)$$

$$H_0(z_i) = -\frac{\partial^2}{\partial z_i^2} + V(z_i), \quad (21)$$

and

$$H'(\lambda_i, \alpha_i) = \frac{2\lambda_i}{\rho_i + \alpha_i} - \frac{2}{\sqrt{\rho_i^2 + (z_i - D)^2}}, \quad (22)$$

where we have introduced two parameters  $\lambda_i$  and  $\alpha_i$  to be determined by the variational principle. Because of the cylindrical symmetry, exact solutions  $\psi_{m_i}(\lambda_i, \alpha_i, \rho_i) e^{im_i \phi_i}$  of

$H_0(\lambda_i, \alpha_i, \rho_i, \phi_i)$  can be obtained by the series expansion method similar to what have been shown above. Using the exact solution  $\psi_{m_i}(\lambda_i, \alpha_i, \rho_i) e^{im_i \phi_i}$  with the lowest eigenfunction  $\psi(z_i)$  of  $H_0(z_i)$  as a trial function, the lowest energy levels of different  $m_i$  states of  $H(i)$  can be found by a variational calculation.

In a similar fashion, we rewrite Eq. (4) for  $D^-$  as

$$H_D = H_0(\lambda_1, \alpha_1, \rho_1, \phi_1) + H_0(z_1) + H_0(\lambda_2, \alpha_2, \rho_2, \phi_2) + H_0(z_2) + H_{int}(\lambda_1, \alpha_1, \lambda_2, \alpha_2) \quad (23)$$

with

$$H_{int}(\lambda_1, \alpha_2, \lambda_2, \alpha_2) = H'(\lambda_1, \alpha_1) + H(\lambda_2, \alpha_2) + \frac{2}{\sqrt{\rho_{12}^2 + (z_1 - z_2)^2}}. \quad (24)$$

Using the eigenfunctions  $\psi(z_i) \psi_{m_i}(\lambda_i, \alpha_i, \rho_i) e^{im_i \phi_i}$ , we can easily construct the trial function  $\Psi_M$  of Chandrasekhar type<sup>20</sup> with the well-defined total magnetic quantum number  $M = m_1 + m_2$ . Thus we have the trial functions

$$\Psi_0 = \psi(z_1) \psi(z_2) \Phi_0 \quad (25)$$

with

$$\Phi_0 = A_1 (1 + c \rho_{12}) \psi_0(\lambda_1, \alpha_1, \rho_1) \psi_0(\lambda_2, \alpha_2, \rho_2) + \psi_0(\lambda_1, \alpha_1, \rho_2) \psi_0(\lambda_2, \alpha_2, \rho_1) \quad (26)$$

for the lowest singlet  $s$ -like state, and

$$\Psi_{-1}^{\pm} = \psi(z_1) \psi(z_2) \Phi_{-1}^{\pm} \quad (27)$$

with

$$\Phi_{-1}^{\pm} = A_2 (1 + c \rho_{12}) \psi_0(\lambda_1, \alpha_1, \rho_1) \psi_{-1}(\lambda_2, \alpha_2, \rho_2) \times e^{-i\varphi_2 \pm \psi_0(\lambda_1, \alpha_1, \rho_2) \psi_{-1}(\lambda_2, \alpha_2, \rho_1) e^{-i\varphi_1}} \quad (28)$$

for the lowest triplet ( $-$ ) and singlet ( $+$ )  $p$ -like states. We have introduced one more variational parameter  $c$  in Eqs. (26) and (28).  $A_1$  and  $A_2$  are normalization constants. Thus the lowest singlet and triplet levels can be obtained by variational calculation.

Let  $E(D^0, 0)$ ,  $E(D^-, 0)$ , and  $E(D^-, -1)$  denote the lowest levels of a donor and the lowest levels of  $D^-$  singlet and triplet states in QD's, respectively. Compared with the definition in quantum wells, the binding energies  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  of the lowest  $D^-$  singlet and triplet states in QD's are, respectively, as follows:

$$E_B(D^-, 0) = 2\gamma + E(D^0, 0) - E(D^-, 0) \quad (29)$$

and

$$E_B(D^-, -1) = 3\gamma - \gamma_b + E(D^0, 0) - E(D^-, -1). \quad (30)$$

We should point out that the binding energies are only dependent on  $\gamma$  since the Zeeman energy in  $E(D^-, -1)$  is also equal to  $-\gamma_b$ . The energy difference  $\Delta E_{ts}$  between the lowest triplet and singlet states is given as

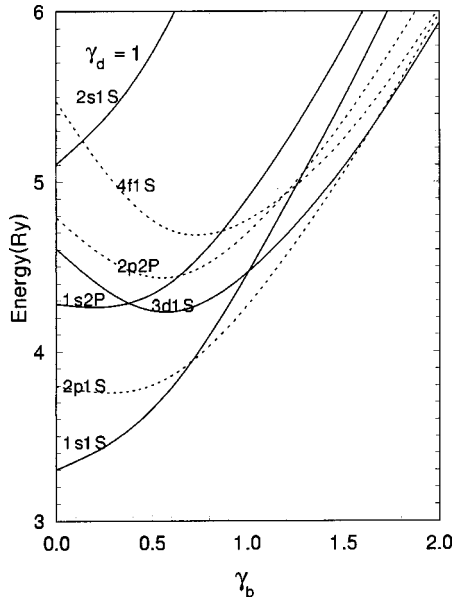


FIG. 1.  $E(n,m;N,M)$  versus  $\gamma_b$  for  $1s1S$ ,  $2s1S$  and the lower  $2p1S$ ,  $1s2P$ ,  $3d1S$ ,  $2p2P$ , and  $4f1S$  states of two electrons in a 2D QD with  $\gamma_d=1$ . The solid and dashed lines represent spin-singlet and spin-triplet states, respectively.

$$\begin{aligned}\Delta E_{ts} &= E(D^-, -1) - E(D^-, 0) \\ &= \gamma - \gamma_b + E_B(D^-, 0) - E_B(D^-, -1).\end{aligned}\quad (31)$$

#### IV. SPIN OSCILLATION

By using the developed model in the previous section, we specify labeling of quantum levels of two electrons in a QD without an ion center. As shown in Eq. (18), the levels  $E(n,m;N,M)$  can be labeled by four symbols  $n$ ,  $m$ ,  $N$ , and  $M$ . The even and odd  $m$  correspond to the spin singlet ( $s=0$ ) and triplet ( $s=1$ ) states, respectively, because of the Pauli exclusion principle. We have states  $1s$ ,  $2p$ ,  $2s$ ,  $3d$ ,  $3p(1S, 2P, 2S, 3D, 3P)$ , and so on if the principal quantum numbers  $n = n + |m| + 1$  ( $N = N + |M| + 1$ ) is used instead of  $n$  ( $N$ ) and the notation  $s, p, d, \dots$  ( $S, P, D, \dots$ ) are used for  $|m|$  ( $|M|$ ) =  $0, 1, 2, \dots$ .

For the sake of clearness, we only plot the lower levels in QD's with  $\gamma_d=1$  as a function of  $\gamma_b$  in Fig. 1. In order to understand the role of the electron-electron interaction in two-electron spectra of QD's in a magnetic field better, we first describe the characteristics of the energy levels of two electrons in QD without interaction. The splitting of energy levels is induced by the Zeeman terms as  $\gamma_b$  increases from zero. The lower levels first decrease and then increase with increasing  $\gamma_b$ . There are minima for the states with negative integers of  $m$ . These results are caused because of the two interactions of the Zeeman term and the parabolic potential in a range of small  $\gamma_b$ , compared with  $\gamma_d$ . However, there is no splitting for the states with  $M=m=0$ , and the levels increase monotonically.

The electron-electron interaction can significantly change the spectra in QD's in a magnetic field. As shown in Fig. 1, the degeneracy of  $1s2P$  and  $2p1S$  states and that of  $3d1S$ ,  $2p2P$ , and  $2s1S$  ones are lifted by the interaction as  $\gamma_b$

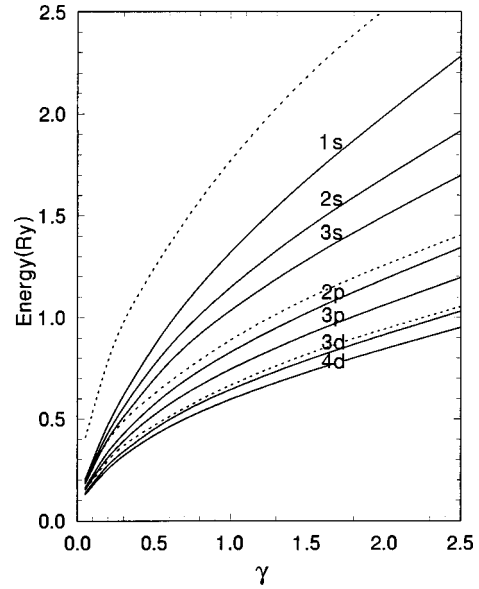


FIG. 2.  $E_r(n,m)$  versus  $\gamma$  for  $1s$ ,  $2s$ ,  $3s$ ,  $2p$ ,  $3p$ ,  $3d$ , and  $4d$  states in the relative-motion system. The solid and dashed lines represent the results obtained by the exact solutions and the first-order perturbation, respectively.

= 0. The level ordering is changed with increasing  $\gamma_b$ , and the intersections between different states appear. An obvious feature of Fig. 1 induced by the interaction is the intersections between the lower levels. It presents the spin-singlet-spin-triplet oscillation of the ground state with  $\gamma_b$ , i.e.,  $1s1S - 2p1S - 3d1S - 4f1S$  states and so on, shown in the figure.

For a better understanding of the singlet-triplet oscillation, it is interesting to study the electron-electron interaction energies  $E_r(n,m)$ , defined by Eq. (17). For a fixed state, it is only dependent on  $\gamma$  so that we plot  $E_r(n,m)$  as a function of  $\gamma$  in Fig. 2. It is readily seen that the  $E_r(n,m)$  increases with  $\gamma$  and the ordering is as follows:  $E_r(0,0) > E_r(1,0) > E_r(2,0) > E_r(0,1) > E_r(1,1) > E_r(0,2) > E_r(1,2) \dots$ . We should point out that the ordering will be changed if the form of confining potential of QD's is changed. It, however, is important to note that for a fixed  $n$ ,  $E_r(n,m)$  are always decreased with increasing  $|m|$  because of the deviation of wave functions from the Coulomb center.

It is accurate enough to calculate the  $E_r(n,m)$  with the use of the first-order perturbation as  $\gamma$  is sufficiently large compared with the electron-electron interaction. Then,  $E_r(n,m)$  are given by

$$E_r(n,m) = \left\langle \phi_{nm}(r) \left| \frac{2}{r} \right| \phi_{nm}(r) \right\rangle, \quad (32)$$

where  $\phi_{nm}(r)$  are normalized radial wave functions of Eq. (12) without the electron-electron interaction term. Using Eq. (32), we can easily find the ordering mentioned above. Furthermore, the values are proportional to  $\gamma^{1/2}$  and always larger than the corresponding ones obtained by the exact solutions. For  $n=0$ , for example, the values given by Eq. (32) is as follows:

$$E_r(0,m) = \begin{cases} (\pi\gamma)^{1/2} & \text{for } m=0, \\ (\pi\gamma)^{1/2} \frac{(2|m|-1)!!}{2|m|!!} & \text{for } |m|>0. \end{cases} \quad (33)$$

Compared with exact values shown in Fig. 1, it is obviously seen that the larger the  $\gamma$  and  $|m|$  are, the less the difference between them is.

On the basis of the above discussion, we explain the oscillation in Fig. 1 as follows. For  $N=n=0$ , the level increases with increasing  $|m|$  as  $\gamma_b=0$ . On the other hand, the levels with  $N=n=0$  and negative integers of  $m$  excluding the interaction, approach the same level as  $\gamma_b \rightarrow \infty$ . For a fixed  $\gamma_b$ , however, the interaction energies decrease with increasing  $|m|$ . It means that the level sequence of  $1s1S-2p1S-3d1S-4f1S$  states and so on is in order of decreasing magnitude as  $\gamma_b \rightarrow \infty$ . This is the reason why the spin-singlet-spin-triplet oscillation with magnetic field appears. It is clear that there is no oscillation without the electron-electron interaction.

Using Eqs. (18) and (33), in fact, the equation for estimating positions of the spin changes can be deduced. The position  $\gamma_{bm}$  for  $m(<0) \rightarrow m-1$  parity change is determined by the following equation:

$$\begin{cases} \frac{1}{2} (\pi\gamma_m)^{1/2} = \gamma_m - \gamma_{bm} & \text{for } m=0, \\ \frac{(2|m|-1)!!}{(2|m|+2)!!} (\pi\gamma_m)^{1/2} = \gamma_m - \gamma_{bm} & \text{for } |m|>0, \end{cases} \quad (34)$$

where  $\gamma_m = (\gamma_d^2 + \gamma_{bm}^2)^{1/2}$ . In general, the prefatory coefficients on the left-hand side of Eq. (34) are less than the corresponding exact ones. Note that the wave functions are different between the two cases, and it can be understood easily.

It is interesting to compare the above spectra with the others in QD's with different sizes to see what kind of difference appears and the important role of electron-electron interaction in various spectra. For the purpose, we have calculated the spectra using  $\gamma_d=0.2$  instead of  $\gamma_d=1$ . First, we should point out that the spectra excluding the interaction are exactly the same as that of  $\gamma_d=1$  except for the different scale. However, the situation is quite different between the two cases after the interaction is included. As  $\gamma_b=0$ , the level ordering is different because of the quantum-size effects.<sup>21</sup> The level ordering and the intersections are also different as  $\gamma_b \neq 0$ . The oscillations appear in the different regions of  $\gamma_b$ . What we have mentioned above means that the electron-electron interaction could play an important part in the spectra and also is one of the effects to make up the magnetic fingerprints of QD's. The other effects may be induced by dot shapes, doping impurities, and so on. The part of impurity effects will be discussed in the next section. To close this section, it is important to point out that the phenomenon of the spin oscillation is independent of the shapes and sizes of QD's while the field dependence of spectra is related to the structures of QD's.

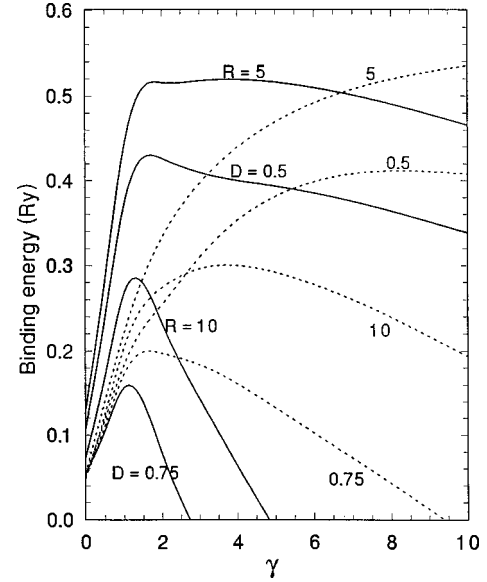


FIG. 3.  $E_B(D^-, 0)$  (solid) and  $E_B(D^-, -1)$  (dashed) versus  $\gamma$  for  $D=0, R=5$ ;  $D=0.5, R=0$ ;  $D=0, R=10$ ; and  $D=0.75, R=0$ , respectively.

## V. $D^-$ BINDING AND OSCILLATION REDUCTION

It is very interesting to study electronic structures in such  $D^-$ -dot broken-symmetry system under a magnetic field since there are two kinds of Coulomb (ion-electron and electron-electron) interactions except the confinement and Zeeman interactions induced by the dot and field. To provide answers to the questions mentioned in Introduction, it is better to study first the interplay of two kinds of Coulomb interactions which are strongly related to the  $D^-$  binding energies in such system.

In two-electron-dot system, only electron-electron Coulomb interaction exists. The spin oscillation appears due to the decreases of the interaction energies with increasing  $|m|$  as shown in Eqs. (33) and (34) and discussed above in Sec. IV. There are both of electron-electron and positive-ion-electron interaction energies in the  $D^-$ -dot system. One is larger than zero and the other less than zero. For the dot-center  $D^-$ , both of the lowest singlet and triplet binding energies increase with magnetic field  $\gamma_b$  and the ratios of  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  to  $E_B(D^0, 0)$  approach constants and freeze at the high-field limit. The former is always larger than the latter so that there is no spin oscillation.

It is quite different for the broken symmetry system, i.e., the off-dot-center  $D^-$  ( $D \neq 0$ ) or  $D^-$  in the graded ( $R \neq 0$ ) dots. In order to show the difference and to better understand the broken symmetry effects on the binding energies and, then, the spin oscillation, we have calculated the  $D^0$  ground states, the lowest singlet and triplet states in the broken symmetry QD's of  $V_0=40$  and  $L=2$  with  $D=0, R=5$  (a),  $D=0.5, R=0$  (b),  $D=0, R=10$  (c), and  $D=0.75, R=0$  (d), respectively.

It is found that the  $E_B(D^0, 0)$  decreases with increasing broken symmetry, i.e.,  $D$  and  $R$  while the  $E_B(D^0, 0)$  always increases with  $\gamma$  no matter what  $R$  and  $D$  are. This is not difficult to understand so that it is not shown here. Figure 3 depicts  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  as a function of  $\gamma$  for different  $R$  and  $D$ . Here it is readily seen that their variation

with  $\gamma$  is not monotonic and quite different from that of  $E_B(D^0, 0)$ . It means that the ratios do not approach constants and there is no frozen-out effect at all. More important is the difference between  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  which is related to the spin oscillation. With increasing  $\gamma$ , as shown in Fig. 3,  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  increase to their maxima and then decrease. However, the values and positions of the maxima which are dependent on  $R$  and  $D$  are quite different between  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  and then the intersections between them appear. The crossing points  $\gamma_c$  are equal to 6.75, 5.4, 1.65, and 0.5 for the (a), (b), (c), and (d) cases, respectively. Furthermore, all of  $E_B(D^-, 0)$  and  $E_B(D^-, -1)$  may become negative when  $\gamma$  is sufficiently large. As shown in the figure, for example, it occurs at  $\gamma = 2.7$  and  $\gamma = 9.3$  for the singlet and triplet states of the QD with  $D = 0.75$ , respectively. Thus a strong magnetic field  $\gamma_b$  can dissociate a  $D^-$  into an electron and a  $D^0$  in the broken symmetry QD.

The phenomena observed in Fig. 3 can be understood qualitatively as follows. The distance between the ion-center and the  $\psi(z_i)$ -maximum point enlarges and for a fixed  $\gamma_b$ , then, the absolute values of ion-electron interaction energies decrease because of the symmetry break due to a nonzero  $R$  or  $D$ . The difference of the ion-electron interaction energies between the symmetry ( $D=R=0$ ) and broken symmetry cases increase with  $\gamma_b$  strongly. However, it is quite different for electron-electron interaction energies. Especially in the strong-field region, the difference of electron-electron interaction energies between the two cases slightly changes with  $\gamma_b$  compared with that of ion-electron ones. This is the reason why there is not any singlet or triplet bound state and  $D^-$  dissociate into an electron and a  $D^0$  in the broken symmetry QD's with sufficiently large magnetic field.

Compared with the lowest triplet states, the lowest singlet states are more sensitive to the broken symmetry,  $D$  and  $R$ . For the broken symmetry case, the decrease of the ion-electron interaction of the singlet states is much larger than that of the triplet states because the singlet states are more localized than the triplet states. The Coulomb and exchange energies of the singlet states increase due to the closeness of the two orbitals while those of the triplet ones change slightly. More important is that the exchange energies enhance the electron-electron interaction energies of two electrons in the singlet states and suppress those in triplet states. What mentioned above is the reason why  $E_B(D^-, -1)$  changes with  $\gamma_b$  smoothly and could be larger than  $E_B(D^-, 0)$ .

Now, we are ready to show and explain the ion effect on the spin oscillation. In Fig. 4, we plot  $\Delta E_{ts}$  as a function of  $\gamma_b$  for different  $D$  or  $R$ . It is clearly seen that  $\Delta E_{ts}$  decreases with increasing  $\gamma_b$  because  $\gamma - \gamma_b$  becomes less. However, the  $\Delta E_{ts}$  is mainly determined by the binding energy difference between the singlet and triplet states as  $\gamma_b$  is large enough and  $\gamma - \gamma_b$  approaches zero. For the broken symmetry case, the  $\Delta E_{ts}$  becomes zero at a magnetic field  $\gamma_0$ , which satisfies the condition  $(\gamma_0^2 + \gamma_d^2)^{1/2} > \gamma_c$ . As shown in Fig. 4, the less  $D$  or  $R$  is, the stronger  $D^-$  states and the larger  $\gamma_0$  are. For the symmetry case ( $D=R=0$ ) with an ion center,  $\Delta E_{ts}$  is always larger than zero. For a fixed  $\gamma_b$ , the less  $D$  or  $R$  is, the larger  $\Delta E_{ts}$  is. What this means is that a

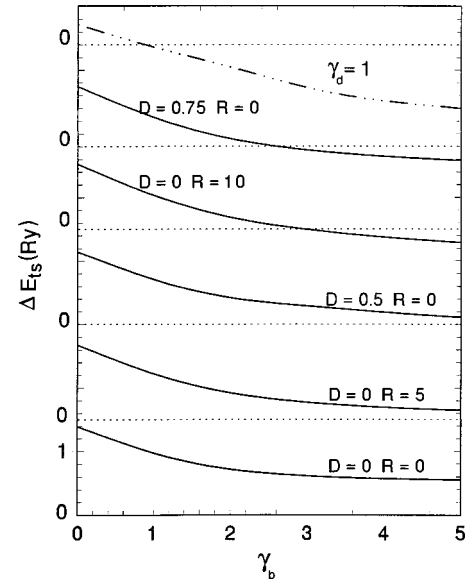


FIG. 4.  $\Delta E_{ts}$  versus  $\gamma_b$  for QD's of  $\gamma_d = 1$  with  $D = 0, R = 0$ ;  $D = 0, R = 5$ ;  $D = 0.5, R = 0$ ;  $D = 0, R = 10$ ; and  $D = 0.75, R = 0$ , respectively. The dot-dashed line represents that of two electrons in the QD of  $\gamma_d = 1$  without the ion center. For the sake of clearness, the  $\gamma_b$  axes (dashed lines) have been shifted up.

positive ion can suppress the spin oscillation of two electrons completely and partly in the symmetry and broken symmetry cases, respectively, and that the oscillation reduction or the starting point  $\gamma_0$  of the oscillation is strongly dependent on  $\gamma_d$ ,  $R$ , and  $D$ . For  $R = 0$  the  $\gamma_0$  approaches its maximum ( $\infty$ ) and minimum as  $D \rightarrow 0$  and  $D \rightarrow \infty$ , respectively. It corresponds to that of two electrons in the QD with and without the ion center, as shown in the figure.

## VI. SUMMARY

The exact spectra of two electrons in quantum dots (QD's) with parabolic potentials in a magnetic field are obtained by using different series solutions in different regions for the radial equation of the relative motion. It has been found that the electron-electron interaction can significantly change the spectra. An obvious feature induced by the interaction is the intersections between the lower levels. It presents the spin oscillation of the ground state with  $\gamma_b$ , i.e.,  $1s1S - 2p1S - 3d1S - 4f1S$  states and so on. The phenomenon is independent on the shapes and sizes of QD's.

The lowest singlet and triplet states of two electrons in the broken symmetry QD's with a positive ion in a magnetic field have been calculated by a variational method. For the broken symmetry case, the ion-electron and electron-electron interactions change with strong magnetic field in different ratios and there is no frozen-out effect at all. Furthermore, exchange energies enhance the electron-electron interactions in singlet states and suppress those in triplet states so that the variation of  $E_B(D^-, 0)$  is quite different from that of  $E_B(D^-, -1)$  and the spin oscillation is influenced. An interesting phenomenon is that a strong magnetic can reduce the binding energies of  $D^-$  states in the broken symmetry QD's until the dissociation. The ion has an important effect on the spin oscillation and the oscillation reduction are strongly dependent on the broken symmetry,  $D$  and  $R$ .

The present results will be useful to understand the optical and transport properties in quantum dots under magnetic field and explaining the experimental phenomena related to ion-electron and electron-electron interactions in QD's. Finally, it can be expected that the proper electronic structures of QD's and the related properties will be obtained if the sizes and shapes of QD's including doping and the numbers of electrons are better controlled. Therefore, it is very important to study electronic structures of few electrons with few ions in QD's with different sizes and shapes.

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